Evidence for a common physical description of non-Fermi-liquid behavior in f-electron systems

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The non-Fermi-liquid (NFL) behavior observed in the low temperature specific heat $C(T)$ and magnetic susceptibility $\chi(T)$ of f-electron systems is analyzed within the context of a recently developed theory based on Griffiths singularities. Measurements of $C(T)$ and $\chi(T)$ in the systems $\mathrm{Th_{1-x}U_xPd_2Al_3}$, $\mathrm{Y_{1-x}U_xPd_3}$, and $\mathrm{UCu_{5-x}M_x}$ (M = Pd, Pt) are found to be consistent with $C(T)/T \propto \chi(T) \propto T^{-1+\lambda}$ predicted by this model with $\lambda < 1$ in the NFL regime. These results suggest that the NFL properties observed in a wide variety of f-electron systems can be described within the context of a common physical picture.

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Transport, thermal, and magnetic measurements on a number of chemically substituted rare earth and actinide compounds have revealed low temperature physical properties that show striking departures from the predictions of Fermi-liquid theory [1]. Several theoretical models have been developed to account for the non-Fermi-liquid (NFL) behavior observed in f-electron materials. These models include a multichannel Kondo effect of magnetic or electric origin [2–4], fluctuations of an order parameter in the vicinity of a second order phase transition at $T = 0$ K [5–7], a disordered distribution of Kondo temperatures [8,9], and an electron polaron model for heavy fermion systems [10]. However, none of these models has been able to account for all of the NFL characteristics observed in the wide variety of systems that belong to this new class of strongly correlated f-electron materials. Three of us (A. H. C. N., G. E. C., and B. A. J.) have recently proposed a model where NFL behavior is associated with the proximity to a quantum critical point and the formation of magnetic clusters in the paramagnetic phase due to the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction in the presence of magnetic anisotropy and

disorder inherent in alloyed materials [11]. This model predicts that various physical properties diverge with decreasing temperature as weak power laws of temperature and that this behavior persists over appreciable ranges of substituent concentration, similar to what has been observed in a number of f-electron materials.

In this letter, we compare low temperature specific heat $C(T)$ and magnetic susceptibility $\chi(T)$ data for the ordered f-electron sublattice systems $\mathrm{UCu_{5-x}M_x}$ (M = Pd, Pt) (Ref. [12,13]) and the disordered f-electron sublattice systems $\mathrm{Th_{1-x}U_xPd_2Al_3}$ (Ref. [14]) and $\mathrm{Y_{1-x}U_xPd_3}$ (Ref. [15]) with the predictions of the new model proposed in Ref. [11]. In particular, we compare fits of our specific heat data with logarithmic functions of temperature, as predicted in several previously developed theories of NFL behavior, to fits with power laws in temperature as suggested by this new model. The data are all derived from our own measurements, some of which are new and reported here for the first time and others of which have been published previously [13–17]. We find that the NFL behavior of $C(T)$ and $\chi(T)$ in these compounds is consistent with the predictions of the model proposed in Ref. [11]. Specifically, $C(T)/T$ and $\chi(T)$ can be described by divergent power laws in temperature at the lowest temperatures for all of the compounds investigated. Our results suggest that this general scenario holds promise for understanding the NFL behavior in this new class of f-electron materials.

Details of the procedures used to prepare the polycrystalline uranium compounds studied in this work are described elsewhere [13,15]. Magnetization $M(T)$ measurements were performed after field cooling a sample to the lowest temperature using a commercial SQUID magnetometer (Quantum Design) in fields of 0.5 and 1 tesla and at temperatures in the range 1.8 K $\leq T \leq$ 300 K. The low temperature $M(T)$ data (0.4 K $\leq T \leq$ 2 K) were acquired with a ³He Faraday magnetometer (FM). Heat capacity measurements were made in a ³He semi-

TABLE I. Exponent λ obtained from fits of Eq. (1) to specific heat (λ_C) and magnetic susceptibility (λ_χ) data for the f-electron systems shown in Figs. 1 and 2.

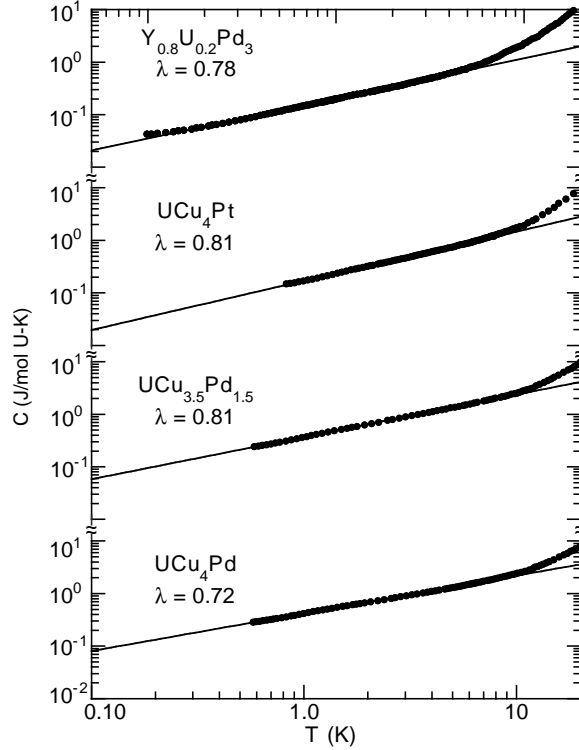|	Th$_{1-x}$U$_x$Pd$_2$Al$_3$						UCu$_{5-x}$Pd$_x$		UCu$_4$Pt	Y$_{0.8}$U$_{0.2}$Pd$_3$
x	0	0.2	0.4	0.6	0.8	0.9	1.0	1.5		
λ_C	1	0.85	0.81	0.84	0.81	1	0.72	0.81	0.83	0.76
λ_χ	0.87	0.6	0.63	0.63	0.6	—	0.72	0.78	0.77	0.70

FIG. 1. Log-log plot of specific heat C vs temperature T for several U-based NFL systems. Solid lines are fits of the data to Eq. 1.
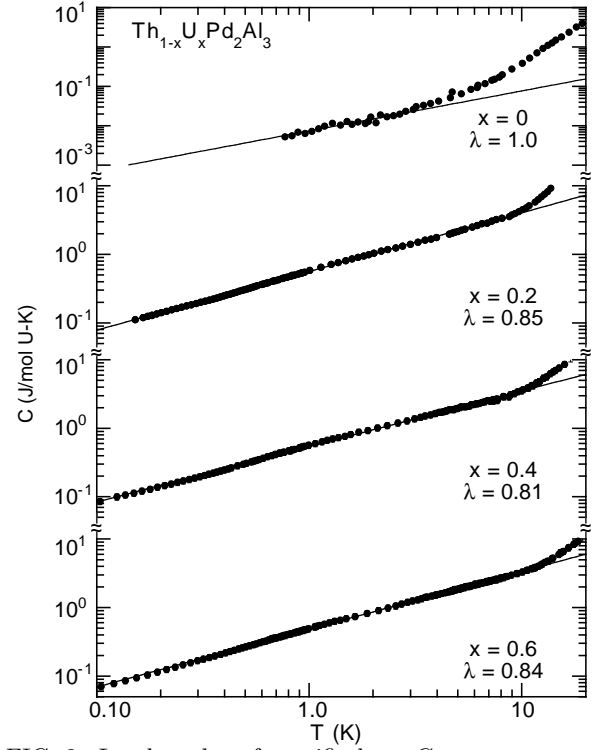

FIG. 2. Log-log plot of specific heat C vs temperature T for the Th$_{1-x}$U$_x$Pd$_2$Al$_3$ system for several values of x. Solid lines are fits of the data to Eq. 1.

adiabatic calorimeter with a standard heat-pulse technique.

Log-log plots of C vs T in the NFL regime are shown in Fig.1 for several samples in the Y$_{1-x}$U$_x$Pd$_3$ (x = 0.2) and UCu$_{5-x}$M$_x$ (M = Pt, x = 1; M = Pd, x = 1, 1.5) systems and in Fig.2 for the Th$_{1-x}$U$_x$Pd$_2$Al$_3$ (x = 0, 0.2, 0.4, 0.6) system. In these figures, the error bars are smaller than the size of the symbols. The solid lines represent least squares fits of the expression relating the specific heat C to the magnetic susceptibility χ, given by

$$C(T)/T \propto \chi(T) \propto T^{-1+\lambda} \qquad (1)$$

at low temperatures, where λ is a parameter determined by the best fit. The values of λ for different compounds and/or different compositions x of the chemical substituent are indicated in Figs.1 and 2 and are given in Table 1. To test the quality of the fits, the reduced chi square, χ_ν^2, was calculated [18] and will be discussed below. The log-log plots of C vs T (Figs.1 and 2) reveal that

a power law with $\lambda < 1$ provides an excellent description of the data for all the curves. For the Th$_{1-x}$U$_x$Pd$_2$Al$_3$ system, the power law describes the data from 0.1 K up to 14 K. In the Y$_{1-x}$U$_x$Pd$_3$ and UCu$_{5-x}$M$_x$ systems, the best fit was achieved for 0.4 K $\leq T \leq$ 5 K. Notice that for ThPd$_2$Al$_3$ we get $\lambda = 1$ as expected for a Fermi liquid.

In order to provide a more direct comparison between power law and logarithmic behavior, plots of C/T vs $\log T$ are shown in Fig.3 for three selected compounds, UCu$_4$Pd, Th$_{0.4}$U$_{0.6}$Pd$_2$Al$_3$, and Y$_{0.8}$U$_{0.2}$Pd$_3$. The upturns in $C(T)/T$ at high temperatures are due to the phonon contributions and Schottky anomalies arising from crystalline electric field splitting of the U 5f Hund's rule ground state multiplet and are excluded from the fitting range. These contributions were not subtracted because they could not be estimated with sufficient accuracy. In this figure, both power law (solid lines) and logarithmic (dashed lines) temperature dependences were fit to the data over the same temperature range, and χ_ν^2
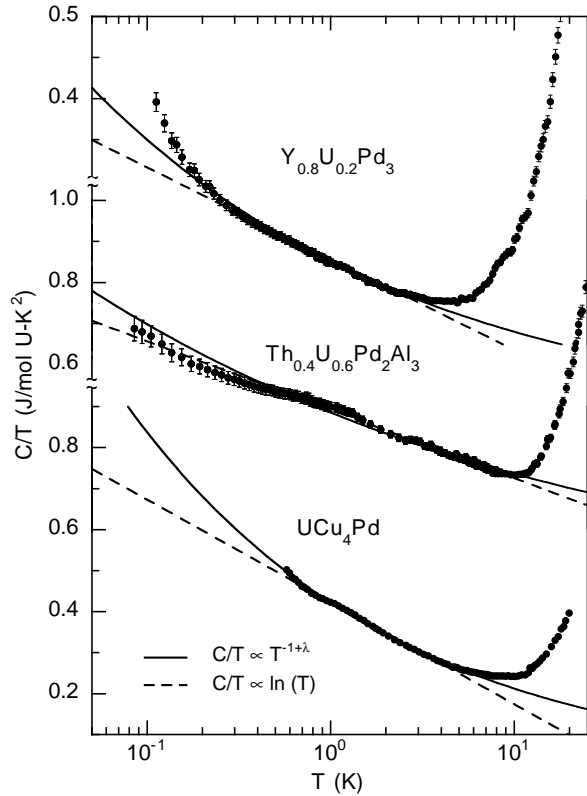
FIG. 3. Semilog plot of specific heat C vs temperature T for selected U-based NFL compounds. Solid and dashed lines are fits to the expressions indicated in the figure.
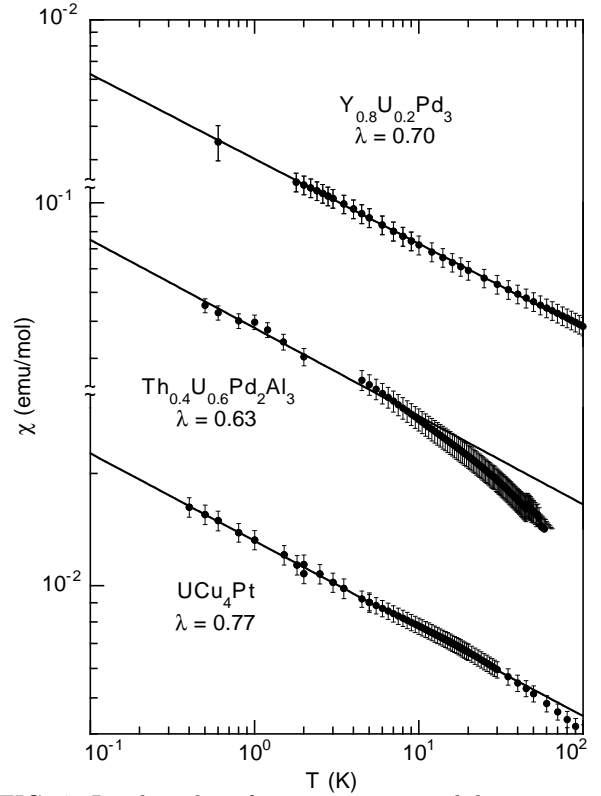

FIG. 4. Log-log plot of magnetic susceptibility χ vs temperature T for selected U-based NFL compounds. Solid lines are fits of the data to Eq. 1.

was then calculated to assess the quality of each fit. Table 2 shows the calculated values of χ_ν^2 for each of the compounds in Fig.3. Recall that for an optimum fit, χ_ν^2 is close to 1 [18]. Although both the logarithmic and power law descriptions of the specific heat agree with the experimental data within the experimental resolution, the reduced chi square indicates that a power law provides a better description of the data than a logarithmic relation over the same temperature range.

Plots of the magnetic susceptibility $\chi(T)$ vs $\ln T$ for UCu_4Pt, $Th_{0.4}U_{0.6}Pd_2Al_3$ and $Y_{0.8}U_{0.2}Pd_3$ are shown in Fig. 4. The data have been fit with power laws (Eq.1) and give values of λ that are close to the values obtained from the power law fits to the $C(T)$ data. Values of λ from these fits are given in Table 1. The magnetic susceptibility was obtained from the linear part of the magnetization M vs magnetic field H isotherm at low fields $\mu H/kT \ll 1$, where μ is the magnetic moment

TABLE II. Calculated values of the reduced chi square χ_ν^2 from fits to specific heat data shown in Fig. 3.

	$Th_{0.4}U_{0.6}Pd_2Al_3$	UCu_4Pd	$Y_{0.8}U_{0.2}Pd_3$
$C/T \propto \ln T$	0.31	1.57	3.48
$C/T \propto T^{-1+\lambda}$	0.94	1.03	0.97

and k is Boltmann's constant. This procedure was especially important at low temperatures where the M vs H curves display negative curvature which is more pronounced at lower temperatures. Assuming the negative curvature of $M(H)$ is intrinsic, this method of extracting $\chi(T)$ suggests that the origin of the NFL behavior is magnetic in nature, since $C(T)/T$ and $\chi(T)$ have the same temperature dependence. We also note that the values of λ obtained from magnetization measurements on the $Th_{1-x}U_xPd_2Al_3$ system reflect the average of the magnetization over crystallites with a hexagonal structure oriented in different directions, which may explain the difference between the values of λ from specific heat and susceptibility in this system. In a single crystal, we expect better agreement between the specific heat and the magnetization [19].

We have shown from our experimental data that the NFL behavior observed in $C(T)$ and $\chi(T)$ given in Eq. 1 is consistent with the existence of Griffiths singularities. The values of λ obtained from the $C(T)$ and $\chi(T)$ data agree with one another within experimental resolution, suggesting that the NFL behavior originates from magnetic interactions. The U-based systems investigated in this work have all of the prerequisites for the Griffiths phase scenario: the Kondo effect, RKKY interactions between the U magnetic mo-

ments, magnetic anisotropy due to spin-orbit interactions, and disorder associated with the chemical substitutions. Furthermore, the imaginary part of the frequency dependent susceptibility $\chi''(\omega)$ of $UCu_{3.5}Pd_{1.5}$ and UCu_4Pd, derived from neutron scattering measurements on these materials [20], is described well by the Griffiths phase result $\chi''(\omega) \propto \omega^{-1+\lambda} \tanh(\omega/T)$ [11] with a value $\lambda \approx 0.7$, in good agreement with the values of λ determined from the above analysis of the $C(T)$ and $\chi(T)$ measurements. NMR and μSR experiments [8] on these same compounds reveal a distribution of susceptibilities given by $\delta\chi/\chi \propto T^{-\lambda/2}$ which is also consistent with the presence of a Griffiths phase at low temperatures [11]. Finally, recent NMR and μSR experiments on $UCu_{5-x}Pd_x$ [8] and pressure experiments on the NFL system $CeRh_2Si_2$ and $CeRh_{2-x}Ru_xSi_2$ [21] indicate that disorder plays an especially important role in NFL behavior. In addition, the relatively large range of substituent concentration x over which the NFL behavior extends in certain NFL systems is consistent with the notion of a Griffiths phase since these systems have rather large values of T_K ($\lesssim 10^2$ K) and the range of x over which the Griffiths phase extends is predicted to be larger for larger values of T_K. Thus, there is sufficient evidence to suggest that the Griffiths phase model is a viable candidate for the NFL properties of the f-electron systems investigated herein. Moreover, recent calculations yield a linear temperature dependence of the resistivity in these systems due to electron scattering by magnetic clusters [22] in agreement with transport data [1].

We point out that in some of these systems it is also possible that a quadrupolar Kondo effect, due to the exchange interaction between the quadrupolar moment of a Γ_3 non-magnetic doublet ground state of U and the conduction electrons, may occur and lead to NFL behavior [16]. However it is not clear that the multichannel Kondo and Kondo disorder models, which are single ion models, are applicable at the relatively large substituent concentrations where NFL behavior is often observed in f-electron materials. Models based on the existence of quantum critical points are known to produce power laws in thermodynamic properties [5]. The NFL behavior can be interpreted as a generic feature in the vicinity of a quantum critical point [7]. However, in the absence of disorder and at zero temperature, the quantum critical regions reduce to points in the phase diagram and are not general enough to explain the broad range of substituent concentrations over which NFL behavior is observed. While each of these models have varying degrees of success in describing the temperature dependence of the physical properties in these materials, none has the general applicability to a wide range of f-electron systems, nor the apparent internal consistency found in our analysis in terms of Griffiths singularities.

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